Magneto-photoluminescence studies of $Zn_{1-x}Mn_xTe$/ZnTe multiple quantum-well and quantum dot structures

Ivan J. Griffin[a], Peter J. Klar[a], Daniel Wolverson[a,*],
J. John Davies[a], Bernard Lunn[b], Duncan E. Ashenford[b],
and Torsten Henning[c]

[a]School of Physics, University of East Anglia,
Norwich NR4 7TJ, UK
[b]Department of Engineering Design and Manufacture,
University of Hull, Hull HU6 7RX, UK
[c]Department of Physics, Göteborg University and
Chalmers University of Technology, SE-41296 Göteborg, Sweden

Abstract

Wide quantum dots were fabricated from multiple quantum well structures based on $Zn_{1-x}Mn_xTe$/ZnTe ($x = 0.076$) dilute magnetic semiconductors and were investigated via photoluminescence (PL) in a magnetic field. Calculations taking into account the strain in the two types of structure enabled the PL transitions to be identified and show that the dominant emission in the MQWs is from heavy-hole (hh) excitons whereas in the quantum dots, the removal of the strain in the barrier layers generates a large biaxial tensile strain in the quantum wells which shifts the light-hole (lh) exciton to lower energy than the hh exciton. The lh exciton σ^+ transition is virtually independent of magnetic field whilst the hh exciton is field-dependent. Thus, at fields of 1 to 2 Tesla, the hh exciton σ^+ transition again becomes the lowest energy transition of the quantum dots. These observations are described by a model with a chemical valence band offset of 30 % for $Zn_{1-x}Mn_xTe$/ZnTe.

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*Corresponding author, Fax: +44 1603 259515, telephone +44 1603 592980, email: d.wolverson@uea.ac.uk

1   Introduction

This paper concerns dots structure fabricated by lithography from dilute magnetic semiconductor (DMS) heterostructures. Although the dots do not have sufficiently small radii for lateral quantum confinement effects to be important, we shall, nevertheless, denote them by the conventional term 'quantum dots'; the importance of studying such dots lies in the need to understand strain effects before attempting to investigate the quantum effects themselves. In earlier work, we employed photomodulated reflectivity (PR) and photoluminescence (PL) to study multiple quantum wells (MQWs) formed from $Zn_{1-x}Mn_xTe/ZnTe$ [1,2] and to investigate the occurence of a field-induced type I-type II transition [3]. Dots fabricated from these MQW structures have also been investigated by PR [4] in the absence of a magnetic field: here we describe the effects on the PL spectra when such a field is introduced.

2   Experimental details

Two $Zn_{1-x}Mn_xTe/ZnTe$ MQWs were grown by MBE on $(1\,0\,0)$-oriented GaSb substrates with $1000\,\text{Å}$ ZnTe buffer layers. Each heterostructure consisted of 10 ZnTe quantum wells of thickness $80\,\text{Å}$ (sample number H561) and $100\,\text{Å}$ (H560) embedded between $Zn_{1-x}Mn_xTe$ barrier layers of $x = 7.6\,\%$ and thickness $150\,\text{Å}$. The samples were nominally undoped [1]. The dot samples were prepared from pieces of the two $Zn_{1-x}Mn_xTe/ZnTe$ MQWs by electron beam lithography for pattern definition followed by Ar^{2+} ion beam etching for transferring the pattern, giving a $3.2 \times 3.2\,\text{mm}^2$ area with a high density of homogeneous tapering pillars of height $\approx 300\,\text{nm}$ (etching stopped before the substrate was reached) and diameter of $\approx 200\,\text{nm}$ [4]. PL experiments were carried out at $1.5\,\text{K}$ in magnetic fields up to 6 Tesla (along the growth axis). Excitation (433 nm) was provided by a UV-pumped Stilbene 3 dye laser and the emitted light was detected in the Faraday geometry.

3   Results and discussion

Figure 1 shows the zero-magnetic field PL spectra of the two $Zn_{1-x}Mn_xTe/ZnTe$ MQW samples and the two corresponding dot samples. We first discuss the free excitons region (above about $2.38\,\text{eV}$). Fig. 1 shows, firstly, that the PL emission from the quantum wells in the dot samples (labelled *e1lh1*) is shifted to lower energy by about 6 meV compared to the corresponding bands (*e1hh1*) in the PL of the MQWs. Secondly, because of stronger confinement in narrower quantum wells, the free exciton emission for the MQW and dot from H561 is at a higher energy than for H560.

The first observation is of particular interest here since it relates to differences in strain. To a first approximation, the MQW samples are strained to the ZnTe lattice constant, resulting in a compressive biaxial strain in the barriers
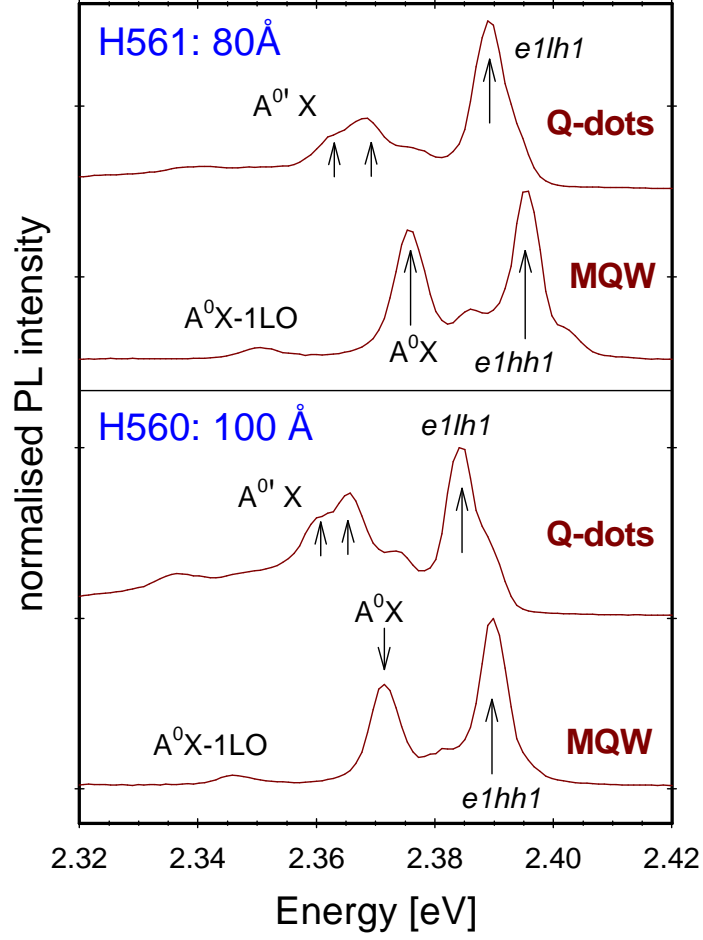
Figure 1: Photoluminescence spectra at $1.5\,\mathrm{K}$ with $433\,\mathrm{nm}$ excitation of the two multiple $\mathrm{Zn}_{1-x}\mathrm{Mn}_x\mathrm{Te/ZnTe}$ quantum well samples (well widths indicated in the figure) and of the quantum dot structures fabricated from them. The spectra are normalised to the same peak height for the dominant transition in each case. The labels of the various peaks are explained in the text.

and zero strain in the quantum wells [1]. In contrast, the dots appear to be free-standing structures with an interatomic spacing in the layer plane intermediate between that of $Zn_{1-x}Mn_xTe$ and ZnTe, there being a consequent tensile biaxial strain in the quantum well layers [2]. For the two cases there is a different ordering of the excitonic energy levels: in the original, unstrained quantum wells, the free exciton emission is expected to have heavy-hole character (confinement effects are smaller for the heavy holes, so that the *e1hh1* is the transition of lowest energy); in contrast, for the dots, there will exist a tensile biaxial strain which may be large enough for the light hole states to become lowest. The experiments described below confirm that this is so.

In the bound state PL (below about $2.38\,eV$), an acceptor-bound exciton line (A^0X) and its first phonon replica (A^0X-1LO) can be seen for both the original MQW structures (Fig. 1). This acceptor is presumed to originate from Sb diffused from the GaSb substrate [5]. The A^0X signal shows no significant magnetic field dependence (see below) and is attributed to the buffer layer (in which the Sb content will presumably be highest). In the PL spectra of the dots, the A^0X band is weaker and an additional acceptor band, $A^{0'}X$, is observed, probably resulting from the nanofabrication process, as reported by other groups [6,7].

The effects of a magnetic field on the PL spectra are shown in Figs. 2 and 3 for the MQW and the dots of H561. The quantum well-related emission bands shift to lower energy with increasing field, whilst the signals attributed to the buffer layer show no discernible shift. We note that the excitonic PL transition σ^+ *e1hh1* (involving the $j_z = -3/2$ heavy hole and the $s_z = -1/2$ electron states) is expected to shift more rapidly with increasing magnetic field than the light-hole like PL transition σ^+ e1lh1 ($j_z = -1/2$, $s_z = +1/2$); this is because, firstly, the field-induced splitting of the heavy hole states is approximately three times bigger than that of the light hole states and, secondly, the magnetic shifts of the single-particle hole and electron states constituting the exciton *add* for the *hh* exciton but have *opposite* sign for the *lh* exciton and approximately cancel. The character of the free exciton emission should therefore change from light to heavy hole-like with increasing field: we shall show that this does indeed occur.

The intensities of the quantum well-related bands (Figs. 2 and 3) change with magnetic field, but this behaviour is hard to interpret, since the magnetic field induces simultaneously changes in the occupation of the states, in the non-excitonic recombination processes and in the degree of confinement. The occurence of a magnetic-field induced type I-type II transition for the σ^+ e1lh1 exciton [3,8] further complicates the situation and we therefore concentrate on the energy changes, rather than the intensity changes, in the PL spectra. Full details of the calculations of the exciton energies are given elsewhere [3] but can be summarised as follows. Five separate steps are carried out for each magnetic field value and for both polarisation states. First, the single-particle potentials for the electrons and the holes are calculated. Strain shifts, interface roughness effects and magnetic field shifts may be included at this stage (we used the model of Stirner et al. [3,9] and assumed ideally smooth interfaces; enhanced paramagnetism effects, which arise because Mn^{2+} ions in the barrier near to
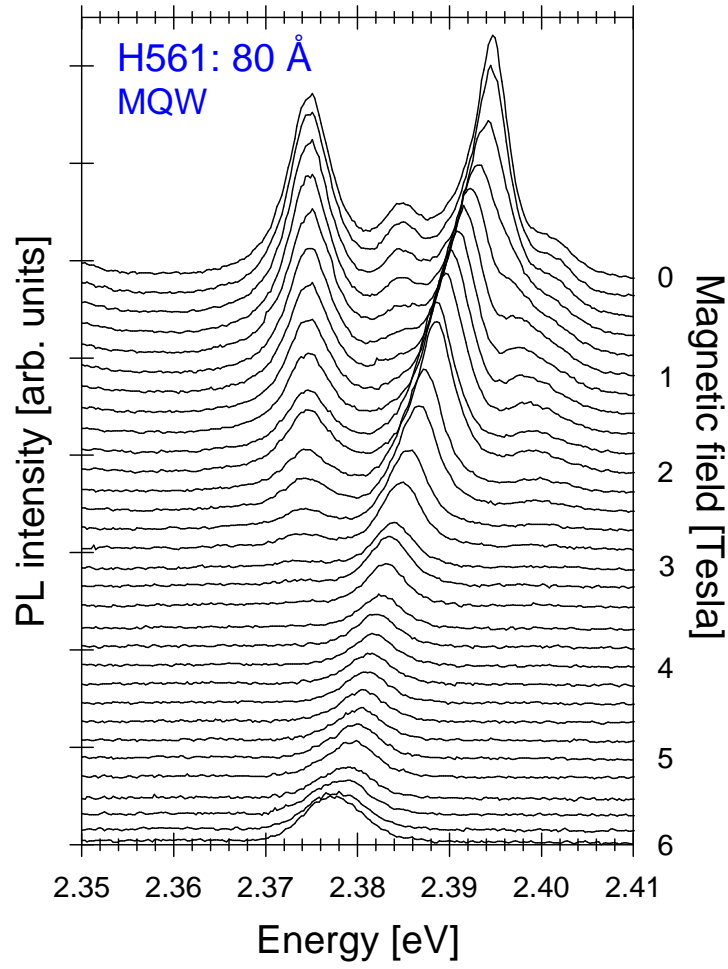
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Figure 2: Photoluminescence spectra of the multiple quantum well sample of well width 80 Å at 1.5 K and in magnetic fields from 0 to 6 Tesla. The spectra are displaced vertically downwards in proportion to the magnetic field and are not normalised. For the identification of the bands at zero magnetic field, see Fig. 1.
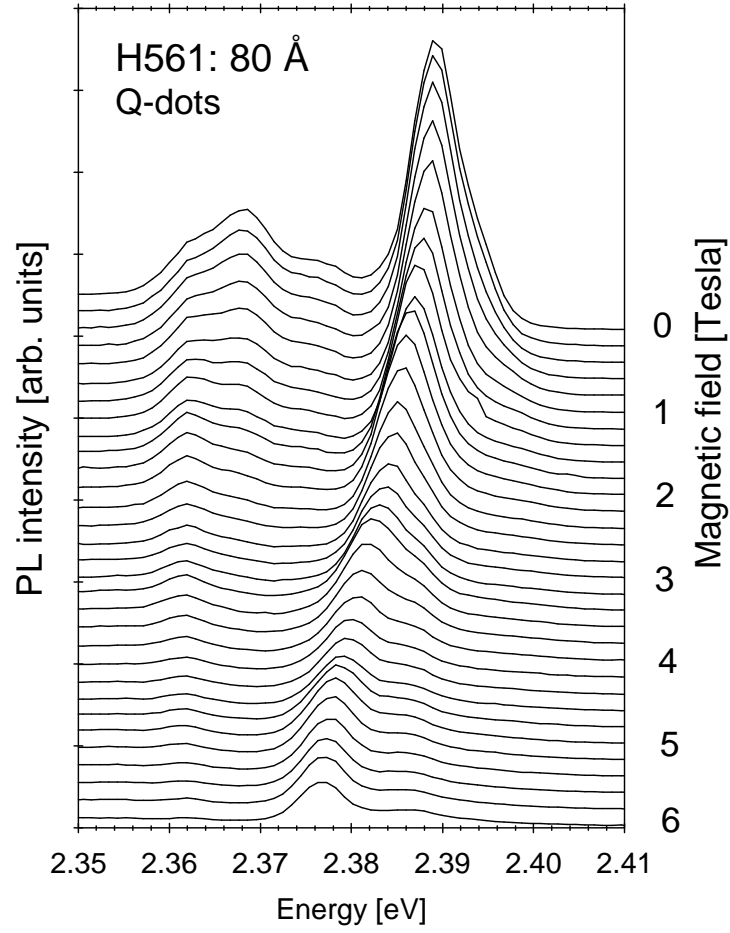
Figure 3: Photoluminescence spectra of the quantum dots fabricated from the multiple quantum well sample of well width 80 Å at 1.5 K and in magnetic fields from 0 to 6 Tesla. The spectra are displaced vertically downwards in proportion to the magnetic field and are not normalised. For the identification of the bands at zero magnetic field, see Fig. 1.

the interface to the quantum well have a lower probability of antiferromagnetic spin-pairing with neighbouring Mn^{2+} ions than do those deep in the barrier, have not been included since such effects assume greatest importance when the Mn^{2+} content of the barrier is large ($> 10\%$) [9]). The remaining steps use a method based on that of Peyla et al. [10] to calculate the excitonic transition energies within the potentials obtained from the first step. The electron single-particle problem is solved and an effective potential for the holes is calculated which includes the QW potential plus that due to the confined electron. The hole energies (and thus the transition energies) are found and an iterative approach used to minimise the exciton energy by variation of the exciton radius parameter in the trial wave function.

The results of these calculations are shown in Fig. 4. We consider first the PL from the MQWs (only the heavy-hole transitions are observed). Using a chemical valence band offset of 30% of the unstrained band gap difference, a barrier Mn concentration of 0.076 and well widths of $100\,\text{Å}$ and $80\,\text{Å}$ respectively with the assumption of smooth interfaces, the calculation yields the solid lines shown in Fig. 4. The agreement with experiment (solid symbols) is good for H561 and poorer for H560: however, the set of parameters used is that which gives the best overall description of the *complete* set of excitonic transitions (both σ^+ and σ^-) in the barriers and wells as observed by modulated photoreflectivity [3].

The same calculation can be applied to the dots, since these are wide enough that no new lateral confinement effects are anticipated. The strain state is modified in the way discussed earlier and leads to the calculated values shown in Fig. 4 (dashed lines). Comparison with experiment shows that, again, the agreement is good for H561 and poorer for H560. However, the main prediction (that in the dots the character of the PL should change from lh- like to hh-like as the magnetic field increases) is borne out in both cases. The near-independence of the lh transition on magnetic field is very clear in Fig. 4 , which shows the crossing of the *lh* and *hh* hole states at a magnetic field of around 1 Tesla.

4   Conclusions

The data obtained from photoreflectivity studies of parent MQW layers were used to predict the excitonic PL energies in dots fabricated from these structures. The only parameter that was changed was the strain: in the dots, the quantum well regions become subject to a large biaxial tensile strain, which causes the lh exciton state to lie below that of the heavy hole, a situation that can then be reversed by application of a magnetic field. The results demonstrate the usefulness of DMS materials in investigations of low-dimensional structures in general and highlight the importance of first establishing the effects of changes in strain before reducing dot diameters sufficiently for lateral quantum confinement to occur.
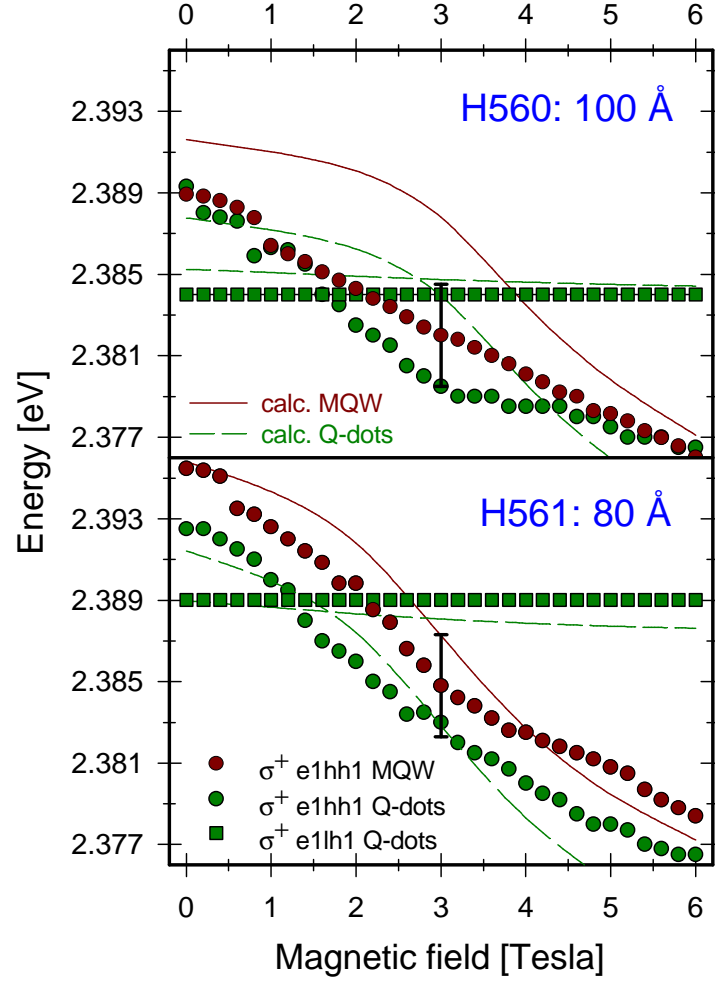
Figure 4: The measured and calculated transition energies of the quantum well excitonic photoluminescence bands as a function of magnetic field for the four structures studied. Solid symbols and solid lines: experimental data and calculations respectively for the multiple quantum wells. Open symbols and dashed lines: experimental data and calculations respectively for the quantum dot structures. The vertical bars indicate the linewidths of the PL bands.

Acknowledgements

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